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**SECOND GENERATION ADVANCED REBURNING  
FOR HIGH EFFICIENCY NO<sub>x</sub> CONTROL**

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## **Abstract**

This project is designed to develop a family of novel NO<sub>x</sub> control technologies, called Second Generation Advanced Reburning (SGAR) which has the potential to achieve 90+ NO<sub>x</sub> control in coal fired boilers at a significantly lower cost than Selective Catalytic Reduction. The ninth reporting period in Phase II (October 1 – December 31, 1999) included preparation of the 10×10<sup>6</sup> Btu/hr Tower Furnace for tests and setting the SGAR model to predict process performance under Tower Furnace conditions. Based on results of previous work, a paper has been prepared and submitted for the presentation at the 28 Symposium (International) on Combustion to be held at the University of Edinburgh, Scotland. A copy of the paper is attached.

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## Executive Summary

This project is designed to develop a family of novel NO<sub>x</sub> control technologies, called Second Generation Advanced Reburning (SGAR) which has the potential to achieve 90+ NO<sub>x</sub> control in coal fired boilers at a significantly lower cost than SCR. The ninth reporting period in Phase II (October 1 – December 31, 1999) included preparation of the 10×10<sup>6</sup> Btu/hr Tower Furnace for tests. Experimental activities included installation of ports for injection of overfire air and N-agent, determination of nozzle droplet distributions at different atomizing conditions, determination of the temperature profile in the furnace, and preparation of CEMs for tests. Modeling activities included calculations of mixing times in the Tower Furnace and estimation of the N-agent droplet evaporation time as function of droplet size. Based on results of previous work, a paper (*Optimization of Advanced Reburning via Modeling*) has been prepared and submitted for the presentation at the 28th Symposium (International) on Combustion to be held at the University of Edinburgh, Scotland. A copy of the paper is attached.

## 1.0 Preparation for $10 \times 10^6$ Btu/hr Proof-of-Concept Tests

Combustion facilities used up to date to obtain data for SGAR optimization and model development included  $0.1 \times 10^6$  Btu/hr Controlled Temperature Tower (CTT) and  $1.0 \times 10^6$  Btu/hr Boiler Simulator Facility (BSF). The experimental portion of the program will continue in a series of proof-of-concept tests at a larger pilot scale facility. The tests are being designed to provide a final indication of the viability of the SGAR technology before proceeding to a full-scale demonstration. The tests will be conducted in EER's Temperature Furnace (TF) at nominally  $10 \times 10^6$  Btu/hr. A three week test series is planned to obtain information on effects of process parameters on  $\text{NO}_x$  reduction. The performance goals in the proof-of-concept tests are to: (1) reduce  $\text{NO}_x$  by up to 95% with net emissions less than  $0.06 \text{ lb NO}_2/10^6 \text{ Btu}$  and (2) minimize other pollutants ( $\text{N}_2\text{O}$ ,  $\text{NH}_3$ , and UBH) to levels lower than reburning and SNCR.

The Tower Furnace (Fig. 1) is a downfired pilot plant combustor with a nominal firing rate of  $10 \times 10^6$  Btu/hr. The facility is designed to provide a large-scale simulation of the flame properties, temperatures, gas compositions, and characteristic mixing times of a coal-fired boiler. As shown in Fig. 1, the Tower Furnace consists of a burner section, radiant furnace, convective pass, and set of air pollution control devices. The burner section can be configured with a single burner or an array of four burners to simulate different types of flames. The facility is equipped with a video camera at the bottom of the furnace, allowing direct monitoring of flame characteristics. The furnace is a refractory lined, water-cooled steel shell. It is square, having dimensions of four feet across and 30 feet in height. It has numerous axial ports, allowing access for injectors and sample probes. The furnace has a turbulent flow field, allowing the impacts of furnace gas mixing and additive entrainment upon process performance to be evaluated. The transition between the furnace and convective pass is a nose section, having geometry and gas flow field characteristics similar to those of a coal fired boiler. Facility air pollution control equipment, which includes a cyclone, baghouse, ESP, and wet scrubber, can be used in varying configurations depending upon test requirements. Because the Tower Furnace provides an accurate simulation of the temperatures, gas compositions, and flow field characteristics of a coal fired boiler, it provides a means of

directly applying results to full-scale systems. Therefore, the Tower Furnace is ideally suited to proof of concept studies before full-scale demonstration of a technology.

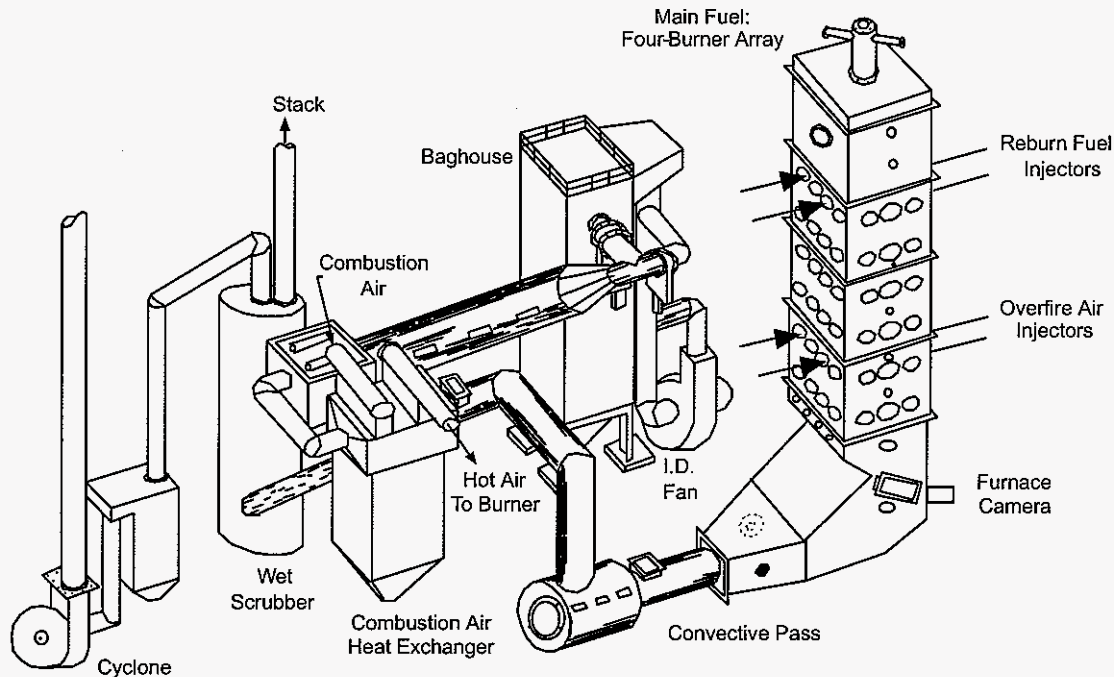


Figure 1. Schematic diagram of the Tower Furnace.

Process performance will be characterized by continuous emissions monitors (CEMs), which provide an online analysis of flue gas composition. The CEMs system consist of a water-cooled sample probe, sample conditioning system (to remove water and particulate), and gas analyzers. Species to be analyzed, detection principles, and detection limits are as follows:

- $O_2$ : paramagnetism, 0.1%
- $NO_x$ : chemiluminescence, 1 ppm
- CO: nondispersive infrared, 1 ppm
- $CO_2$ : nondispersive infrared, 0.1%
- $SO_2$ : nondispersive ultraviolet, 1 ppm
- $N_2O$ : nondispersive infrared, 1 ppm
- $NH_3$ : SCAQMD Method 207 (sampling, Nessler reagent, colorimetry), 1 ppm
- HCN: sampling, ion-specific electrode, 1 ppm

Experimental activities during reported period included installation of ports for injection of overfire air and N-agent, determination of nozzle droplet distributions at different atomizing conditions, determination of the temperature profile in the furnace, and preparation of CEMs for tests. The CEMs system was calibrated for the test program.

## **2.0 Process Modeling**

Modeling activities included calculation of mixing times in the Tower Furnace (for flows of overfire air with N-agent and the flue gas) and estimation of the N-agent droplet evaporation times as function of droplet size. These parameters were calculated and included in the SGAR chemistry-mixing model developed in previous work. The model was setup for calculation of process performance in the Tower Furnace. Modeling calculations have been conducted to assist in test matrix preparation. Modeling and test results will be included in the next quarterly report.

Based on results of previous work, a paper has been prepared and submitted for the presentation at the 28th Symposium (International) on Combustion to be held at the University of Edinburgh, Scotland. The paper entitled *OPTIMIZATION OF ADVANCED REBURNING VIA MODELING* describes the process model that combines a detailed chemical mechanism with a simplified representation of mixing. The model takes into account mixing and thermal characteristics of the  $1.0 \times 10^6$  Btu/hr Boiler Simulator Facility which was used in previous experimental studies to support model development. Modeling suggests that the efficiency of the process strongly depends on amounts of the reburning fuel and N-agent, flue gas temperature at the point of overfire air/N-agent injection, and N-agent evaporation time. The model describes the most important features of AR-Lean which is one of most promising SGAR variants. Application of the model for optimization of AR-Lean performance in the test facility is demonstrated. A copy of the paper is attached to this report.



### **3.0 Future Work**

Future experimental activities will include SGAR tests in the Tower Furnace. Parameters to be varied will include primary fuel (natural gas and coal), reburning zone stoichiometry, N-agent flow rate and injection temperature, and mixing time of overfire air with flue gas. Impacts of mixing upon performance will be evaluated by utilizing two- and four-port configurations for injection of overfire air. Maximum achievable  $\text{NO}_x$  reduction and additive requirements to achieve 95%  $\text{NO}_x$  reduction will be defined. Results will be compared with data from the CTT and BSF to determine the effects of scale upon performance. After the Tower Furnace test, results will be reduced, analyzed, and used in conjunction with the process models to estimate performance in a full-scale boiler.

**Attachment**

**OPTIMIZATION OF ADVANCED REBURNING VIA MODELING**

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## OPTIMIZATION OF ADVANCED REBURNING VIA MODELING

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### Abstract

The Advanced Reburning process (AR) is an integration of basic reburning and N-agent injection. The efficiency of AR depends on many factors, including the amount of the reburning fuel, the amount and location of N-agent injection, and spray characteristics. These parameters can be optimized via computer modeling. This paper describes a model that combines a detailed chemical mechanism with a simplified representation of mixing to describe the AR-Lean process, a combination of basic reburning and N-agent co-injection with overfire air (OFA). The model takes into account mixing and thermal characteristics of a 300 kW combustion facility which was used for experimental studies in support of model development. Modeling suggests that the efficiency of AR-Lean strongly depend on amounts of the reburning fuel and N-agent, flue gas temperature at the point of OFA/N-agent injection, and N-agent evaporation time. The model describes the most important features of AR-Lean. Application of the model for optimization of AR-Lean performance in the test facility is demonstrated.

### Introduction

A family of Advanced Reburning (AR) technologies, an integration of basic reburning and injection of a nitrogen agent (N-agent), is currently under development [1]. These technologies provide  $\text{NO}_x$  control levels similar to that of Selective Catalytic Reduction at lower cost. In AR systems, the N-agent (typically ammonia or urea) can be injected either into the reburning zone (AR-Rich), along with overfire air (AR-Lean), or

downstream in the burnout zone (Reburning+SNCR). AR-Lean is the most commercially attractive option because the N-agent is injected along with the OFA and thus does not require installation of additional ports.

Recent pilot scale experimental data [1-3] demonstrate that different AR systems can provide over 90% NO<sub>x</sub> reduction during natural gas and coal combustion. Installation of each AR technology on a boiler will require design expertise to determine the most efficient process parameters, such as the amount of the reburning fuel, the amount and location of N-agent injection, spray characteristics, etc. Since the efficiency of AR depends on many factors, optimum performance can be achieved if the effects of these factors on process performance are understood. The most efficient approach to AR optimization is to explore the effects of different parameters on NO<sub>x</sub> reduction via kinetic modeling, use the model as a guide for selecting the most effective test conditions, and then optimize performance in pilot- and full-scale combustion facilities. Thus, the kinetic model is an important tool in the development of AR technologies.

Kinetic modeling was successfully used to study AR-Rich [2,3] and Reburning+SNCR [4-8] processes. Modeling of the AR-Lean process [6-8] mostly concentrated on the effect of CO coming from the reburning zone on NO<sub>x</sub> reduction by the N-agent. In experimental and modeling studies, Chen et al. [6] showed that the efficiency of the N-agent could be improved by allowing controllable amounts of CO from the reburning zone to enter the burnout zone. The temperature range over which N-agent is effective is also wider in the presence of CO. However, the optimum temperature for N-agent injection (1100 K) was found to be significantly lower than that used in full-scale boilers for OFA injection (1400-1600 K). Zamansky et al. [7] pointed out that at

proper conditions CO oxidation provides additional radicals which partially oxidize the N-agent and improve its efficiency. Alzueta et al. [8] demonstrated that high concentrations of CO coming from the reburning zone significantly decrease the optimum temperature for NO<sub>x</sub> reduction. However, this modeling [8] was done under the assumption of instantaneous mixing. It was also assumed that O<sub>2</sub> was present in the flue gas at the moment of N-agent injection. This assumption is more relevant to the Reburning+SNCR process than to AR-Lean since delayed mixing of OFA and N-agent with flue gas can significantly affect the efficiency of the AR-Lean process.

Thus a realistic chemistry-mixing modeling study of AR-Lean is needed to predict process performance over a wide range of initial conditions, particularly in the temperature range of OFA/N-agent injection utilized in full-scale boilers. Parameters affecting the efficiency of AR-Lean should be identified and optimized. Specifically, conditions should be determined that would allow injection of N-agent in the temperature range of 1400-1600 K without sacrificing efficiency of NO<sub>x</sub> reduction.

Previous work [9] on basic reburning demonstrated that modeling based on a detailed chemical mechanism with a simplified representation of mixing can be used not only to explore the chemistry of the reburning process, but to identify ranges of process parameters that give optimum process performance. In the current work the model [9] of basic reburning is further developed to describe the effect of N-agent co-injection with OFA on NO<sub>x</sub> reduction. The model is used to (1) identify ranges of parameters that result in the highest AR-Lean efficiency, (2) predict maximum achievable levels of NO<sub>x</sub> reduction, and (3) optimize the process in an experimental combustor.

## Model Setup

The model treats the AR-Lean process as series of four plug-flow reactors. Each reactor describes one of the physical and chemical processes occurring in a boiler: addition of the reburning fuel, NO<sub>x</sub> reduction as a result of the reaction with the reburning fuel, addition of OFA and N-agent, and NO<sub>x</sub> reduction by N-agent and oxidation of partially oxidized products in the burnout zone. The AR-Lean modeling setup is similar to that used for basic reburning [9] except for the injection of N-agent in the third reactor.

The chemical kinetic code ODF [10], for “One Dimensional Flame” was employed to model experimental data. ODF contains the same basic capabilities as Chemkin-II [11], including the evaluation of pressure-dependent and reversible Arrhenius rate expressions, and the specification of time-dependent profiles of temperature and pressure. The solution algorithm has also been formulated to allow for the introduction of an arbitrary profile of heat and/or mass fluxes along the length of the reactor. This capability has proven to be significant for accurate modeling of many types of combustion systems, including reburning with natural gas [9].

The kinetic mechanism [12] includes 447 reactions of 65 C-H-O-N chemical species. The reburning fuel was injected into flue gas at 1670 K. The initial amount of NO was 600 ppm. The temperature of flue gas along the reactor decreased at a linear rate -300 K/s. Variations in the temperature gradient within  $\pm 50^\circ$  showed little effect on modeling predictions.

The mixing process in the reburning zone was described in the model by addition of flue gas to the stream of natural gas during a specified mixing time (so-called inverse mixing). Inverse mixing was also used to describe injection of OFA/N-agent into flue

gas. This approach was found [9] to give a more accurate description of mixing between flue gas and a jet than a traditional description (reactants mixed into flue gas).

Mixing times in the reburning and OFA zones are important parameters that affect efficiencies of the reburning fuel and N-agent. If N-agent is injected in a liquid form, evaporation time of the agent is also important parameter. Values of mixing parameters are specific for each combustion facility and may widely vary. Experimental data used for the model development and validation were obtained in a 300 kW Boiler Simulator Facility (BSF) described elsewhere [9]. Urea was co-injected with OFA perpendicular to the flow of flue gas. Average droplet size was 100  $\mu\text{m}$ . Natural gas was used as main and reburning fuels. These data, injector parameters, and BSF configuration were used to characterize the mixing process in the reburning and burnout zones, and to determine droplet evaporation time of N-agent.

#### *Mixing Time in the Reburning and OFA Zones*

Mixing time and temperature profile in mixing regions were estimated using a single jet in crossflow model, JICFIS [13], for BSF conditions. Major inputs for the model included the velocity and density ratios of the crossflow to the jet, their relative orientation in two dimensional rectangular coordinates, and the initial conditions (diameter, velocity, and temperature) of the jet. The mixing time is determined by integrating the entrainment rate of fluid from the crossflow into the jet. After the point where the entering flow rate equaled the main flow rate, complete mixing was assumed. The mixing time of the reburning jet with flue gas was estimated to be 120 ms. It was assumed that variation in the amount of the reburning fuel had little effect on mixing

time, since the reburning jet in experiments consisted predominately of a nitrogen carrier stream that was held constant.

OFA injection was handled in the same manner as the reburning fuel. For the OFA jet, the mixing time was calculated to be 110 ms. This time is approximate since the actual geometry of injectors does not precisely match the single jet treated by the JICFIS algorithm. However, the results reflect the magnitude of the mixing rate based on the general scale of the problem.

Modeling shows that the value of the mixing time has strong effect on the predicted efficiency of the AR-Lean process as mixing time increases from 0 (instantaneous mixing) to 100 ms. Further increase in mixing time has relatively small effect on  $\text{NO}_x$  reduction. Variation of mixing time from 100 to 140 ms showed little effect on modeling results. Based on the approximations in the JICFIS model, a single mixing time of 120 ms was used for all injections.

#### *Evaporation Time of N-Agent*

Droplet evaporation time can be considered an independent parameter. However, for a selected set of conditions it can depend on other spray parameters such as the flow rate of the liquid and atomization gas.

Droplet evaporation times were predicted using the Computational Fluid Dynamics (CFD) code FLUENT [14]. Monosized water spray droplets were injected into a 1 m/s gaseous plug flow with 10% inlet turbulence, employing slip walls for minimal geometry dependence. The gas was methane combustion products at a stoichiometric



ratio of 1.21. Baseline injection conditions were 100  $\mu\text{m}$ , 300 K droplets injected at 100 m/s co-flow along the duct centerline into 1400 K flue gas.

Modeling shows that droplet evaporation rate is relatively insensitive to the following range conditions, staying in the range of 30-60 ms:

- The temperature difference between gas and spray, in a range of +/- 200 K.
- The initial injection direction relative to the main stream, from 0 to 180 degrees. This does have a significant impact, though, on the streamwise distance required for evaporation (minimized near 90 degrees), which has practical design implications.
- Initial droplet velocity from 50 to 300 m/s. As droplet velocity is decreased below 50 m/s, however, behavior is dominated by rapid deceleration to the main stream velocity, with longer evaporation times.

Figure 1 shows the effect of varying droplet diameter only, from 1 to 300 microns, on evaporation time. Droplet size has a significant impact on evaporation time, and represents a potential control variable for tailoring the distribution of mass evaporation.

Based on these predictions, evaporation time for 100  $\mu\text{m}$  droplets (representative of BSF experiments) is estimated at 45 ms. This is significantly less than estimated BSF mixing times between OFA and flue gas (120 ms). Therefore, comparison of modeling predictions with experimental data assumed instantaneous N-agent evaporation. The predicted effect of droplet evaporation time on AR-Lean efficiency is considered in section *Slow Evaporation of N-Agent*.

### Validation of the AR-Lean Model

Previous work [9] showed that the model of basic reburning could predict the main trends of the process. That model correctly describes  $\text{NO}_x$  reduction efficiencies determined in experiments as functions of the initial  $\text{NO}_x$  concentration, the amount of reburning fuel and the OFA injection temperature. The current AR-Lean model was validated against BSF experiments on reburning with co-injection of urea and OFA.

Figures 2 and 3 show typical comparisons of modeling predictions with experimental data at Nitrogen Stoichiometric Ratio ( $\text{NSR}$ )=1.5 ( $\text{NSR}=[\text{Urea}]/[\text{NO}]_i$ ), where  $[\text{NO}]_i$  is defined as the amount of  $\text{NO}_x$  at the time of OFA/N-agent injection.

Modeling describes the main experimentally observed features of the AR-Lean process. At small reburning fuel heat inputs, the dependence of process efficiency on the OFA/N-agent injection temperature (Fig. 2) is similar to that of SNCR. Modeling accurately predicts the maximum efficiency for 2% reburning, while it underpredicts and overpredicts efficiencies at temperatures lower and higher than the optimum, respectively. At 10% reburning, the optimum in process performance occurs at 1100-1150 K (too low for industrial applications due to poor burnout and ammonia slip).

Figure 3 demonstrates good agreement between modeling predictions and experimental data for basic reburning and AR-Lean processes at an OFA injection temperature of 1300 K (this temperature was found experimentally to give the highest AR-Lean efficiency).  $\text{NO}_x$  reduction in basic reburning increases as the amount of the reburning fuel increases and reaches about 40% at 10% reburning fuel. In AR-Lean the efficiency of  $\text{NO}_x$  reduction is 90-95% for 0- 6% reburning fuel and is insensitive to the amount of reburning fuel.  $\text{NO}_x$  reduction for these conditions is high due to rapid NO/N-

agent mixing in the BSF. As the amount of the reburning fuel increases, the efficiency of the process decreases. This data suggests that AR-Lean practically does not provide advantage over SNCR in combustion facilities with effective mixing of N-agent with flue gas. As discussed below, due to less effective mixing, NO<sub>x</sub> reduction efficiency in AR-Lean in industrial applications can be higher than that of SNCR.

Comparison of modeling predictions with experimental data for other conditions also demonstrates that the model of AR-Lean gives a realistic description of test data. This implies that the mixing and kinetic submodels adequately describe these processes and the model can be used to study the effects of different parameters on trends in the process performance.

### **Parametric Study and Optimization of the AR-Lean Process**

The model of the AR-Lean process incorporates some features that are specific to the BSF. For example, mixing time in the reburning and OFA zones were estimated using characteristics of nozzles utilized in the BSF. Modeling also took into account the temperature profile measured in the BSF. Other combustion facilities have different thermal and mixing characteristics, and this may result in different optimum conditions for AR-Lean. However, differences in process characteristics can be taken into account by adjusting appropriate parameters in the model to optimize the AR-Lean process for a specific facility.

The following parameters were varied in modeling:

- The amount of reburning fuel (0-18% of the total fuel heat input).
- Temperature of flue gas at which OFA and N-agent are injected (1200-1650 K).

- Initial temperature of OFA and N-agent (300-600 K).
- Evaporation time of aqueous N-agent (urea, 0-0.8 s).
- The amount of N-agent (NSR=0-3.0).

Modeling predicted that optimum selection of parameters could result in efficiency of the AR-Lean process in the BSF as high as 95%. The initial temperature of OFA/N-agent was excluded from consideration since preheating of OFA and N-agent results in degradation of the AR-Lean performance and minimum available initial temperature of OFA provided the best performance. The range NSR=1-1.5 was identified as the most effective for the BSF conditions. Assuming that the amount of N-agent is in this range (for example, NSR=1.5), the remaining parameters of interest are the amount of reburning fuel, temperature of flue gas at the point of OFA/N-agent injection and evaporation time of N-agent.

Experimental data on AR-Lean that were used for the model development were obtained at constant N-agent spray characteristics. Thus, N-agent evaporation time was not a test variable. To enable comparison of model predictions with experimental data, the optimization of AR-Lean was limited to two parameters only: the amount of reburning fuel and temperature of flue gas at the point of OFA/N-agent injection. It was assumed in modeling that evaporation of N-agent was fast and occurred within the time scale of the mixing process in the OFA zone. This assumption was made based on estimation of droplet evaporation time for typical BSF conditions. The effect of N-agent evaporation time on  $\text{NO}_x$  reduction is considered in a later section.

### *Fast Evaporation of N-Agent*

At NSR=1.5 a series of modeling runs were conducted to determine the effects of the amount of reburning fuel and the temperature of flue gas at the point of OFA/N-agent injection on AR-Lean  $\text{NO}_x$  reduction (Fig. 4a). The amount of reburning fuel varied from 0 to 10% of the total heat input. For each amount of reburning fuel, the OFA/N-agent injection temperature varied from 1200 K to 1650 K. The experimental data are shown in Fig. 4a as symbols. Comparison of modeling predictions with experimental data shows agreement for a wide range of conditions. The first region with high  $\text{NO}_x$  reductions identified by modeling corresponds to the amount of the reburning fuel in the range 0-6%, and OFA/N-agent injection temperatures about 1280-1350 K. Modeling suggests (and is confirmed by experiments) that the efficiency of  $\text{NO}_x$  reduction in this region is about 90-95%.

As the amount of reburning fuel increases over 6%, the amount of CO coming from the reburning zone becomes significant. Since the optimum temperature range for reaction of N-agent and NO in the presence of CO shifts toward lower temperatures, an increase in AR-Lean performance occurs at higher than 6% reburning fuel for OFA/N-agent injection temperatures less than 1300 K (the second region of high  $\text{NO}_x$  reduction). At 10% reburning, the optimum OFA/N-agent injection temperatures are lower than 1200 K.

Modeling predicts (and experiments confirm) that due to effective mixing, the efficiency of the SNCR process in BSF at 1300 K and NSR=1.5 is very high (over 90%  $\text{NO}_x$  reduction, Figs. 3 and 4a). Therefore, increasing the amount of reburning fuel up to 6% does not significantly improve  $\text{NO}_x$  reduction. However, in full-scale installations,

non-uniformity of the temperature profile, difficulties in mixing the N-agent across the full boiler cross section, and limited residence time for reactions limit effectiveness of SNCR to 30-50%. Some amount of N-agent passes through the system and appears as ammonia slip. Under such mixing conditions, the efficiency of the AR-Lean process may depend more strongly on the amount of reburning fuel. One way to simulate poor mixing of N-agent with flue gas is to reduce the amount of N-agent to the level that provides 40-50%  $\text{NO}_x$  reduction, reflecting the N-agent available to react. Thus, it is of practical interest to study AR-Lean at NSR less than 1.5.

Figure 4b shows performance of the AR-Lean process at NSR=0.7. The maximum  $\text{NO}_x$  reduction in the SNCR process (no reburning fuel), predicted by modeling, is 54%. Modeling results show that at 1300 K (close to optimum temperature for OFA/N-agent injection) the efficiency of AR-Lean process first increases as the amount of the reburning fuel increases, and then decreases. The maximum  $\text{NO}_x$  reduction predicted by modeling is 62%, which is achieved at 5% reburning and is about 8 percentage points higher than the efficiency of SNCR under similar conditions.

Based on modeling predictions, a series of tests were conducted in the BSF to determine the effect of the amount of reburning fuel on  $\text{NO}_x$  reduction in AR-Lean. Test results are shown in Fig. 4b as symbols. Tests confirmed that maximum  $\text{NO}_x$  reduction at NSR=0.7 and 1300 K is achieved around 5% reburning fuel. Maximum reduction observed in tests was 66% - slightly higher than that predicted by modeling.

Thus when droplet evaporation time is smaller than mixing time of OFA in the burnout zone (N-agent is injected as a gas or as small droplets), the AR-Lean process is

most efficient at about 5% reburning fuel and OFA/N-agent injection temperatures in the range of 1280-1350 K.

The second region of high  $\text{NO}_x$  reduction identified by modeling for  $\text{NSR}=0.7$  is located at 10% reburning fuel and an OFA/N-agent injection temperature of about 1200 K. Since in full-scale boilers OFA is usually injected at temperatures higher than 1200 K to achieve full burnout, this result can be considered as being mostly of theoretical interest. The occurrence of high  $\text{NO}_x$  reduction at relatively large reburning heat inputs and low OFA/N-agent injection temperatures is due to the fact that CO formed in the reburning zone interacts with  $\text{NO}_x$ /N-agent chemistry in the OFA zone. As a result, the optimum conditions for  $\text{NO}_x$  reduction are shifted toward lower temperatures.

#### *Slow Evaporation of N-Agent*

To reduce the influence of CO on  $\text{NO}_x$  reduction at large reburning fuel heat inputs, the N-agent can be injected with a delay (as it is done in Reburning+SNCR), or injection can be arranged in such a way that the release of N-agent into the gas phase occurs over a longer period of time. The latter can be done, for example, by injecting larger droplets of aqueous solution containing N-agent. Because of the long time required for large droplets to evaporate and mix with flue gas, N-agent will be delivered to the flue gas with some delay. Both approaches result in N-agent entering flue gas after the OFA and flue gas are already mixed and thus allow for most of the CO to be oxidized before N-agent reacts with  $\text{NO}_x$ . Modeling suggests that utilization of larger droplets increases the efficiency of the AR-Lean process at large reburning fuel heat inputs, but does not affect efficiency of the process at small heat inputs.

Figure 5 shows the predicted performance of AR-Lean as function of the amount of reburning fuel and evaporation time of N-agent at NSR=0.7. Temperature of flue gas at the point of OFA/N-agent injection is optimized with respect to  $\text{NO}_x$  reduction. Optimum temperatures increase from 1300 K at instantaneous evaporation of N-agent to 1500 K at droplet evaporation times close to 800 ms. Modeling predicts that injection of larger droplets of N-agent and utilization of larger amounts of the reburning fuel result in higher levels of  $\text{NO}_x$  reduction. Figure 5 demonstrates that combining of 18% reburning with N-agent injection results in about 80%  $\text{NO}_x$  reduction when droplets with an evaporation time of 100 ms or higher are used, while 5% reburning provides no more than 60%  $\text{NO}_x$  reduction at any droplet evaporation time. Figure 1 demonstrates that droplets larger than about 170  $\mu\text{m}$  provide evaporation times longer than 100 ms.

Injection of larger droplets of N-agent along with OFA is the equivalent of combining reburning with SNCR. Thus, performances of AR-Lean and Reburning+SNCR at optimum conditions should be similar. Note, however, that AR-Lean is much more attractive than Reburning+SNCR from a practical standpoint since no additional N-agent ports are required and OFA serves as the N-agent carrier (no flue gas recirculation required). The model of AR-Lean developed in this work can be used to model the Reburning+SNCR process as well: it only requires introduction of an additional zone that describes mixing of N-agent with flue gas after OFA is added. Figure 6 compares predicted performances of basic reburning, AR-Lean and Reburning+SNCR at conditions (temperatures of flue gas at the point of N-agent injection and droplet evaporation times) that result in the highest optimized level of  $\text{NO}_x$  reduction. Figure 6 also shows AR-Lean performance for injection of small droplets of N-agent (non-



optimized AR-Lean). AR-Lean and Reburning+SNCR result in a significant increase in  $\text{NO}_x$  reduction in comparison with basic reburning. Figure 6 shows that by adjusting the N-agent injection temperature and droplet evaporation time, the efficiency of AR-Lean can be as high as the efficiency of Reburning+SNCR. Figure 6 also demonstrates the importance of optimizing droplet evaporation time in AR-Lean to achieve higher  $\text{NO}_x$  reduction at larger heat inputs of the reburning fuel.

### **Conclusions**

The model developed in this work describes major trends of AR-Lean and can be used for process optimization. Mixing and thermal parameters in the model can be adjusted depending on characteristics of the combustion facility. Modeling identified the following AR-Lean parameters as being most important: amounts of the reburning fuel and N-agent, temperature of flue gas at the point of OFA/N-agent injection, and evaporation time of the N-agent. For evaporation times shorter than the OFA mixing time, AR-Lean is most effective at 3-6% reburning. For evaporation times longer than OFA mixing time, the efficiency of AR-Lean increases as the amount of the reburning fuel increases. The maximum predicted  $\text{NO}_x$  reduction for 18% reburning fuel and  $\text{NSR}=0.7$  is about 80%. Predicted efficiency of  $\text{NO}_x$  reduction increases as the amount of N-agent increases.

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## References

1. Zamansky, V.M., Maly, P.M. and Ho, L. *International Joint Power Generation Conference*, Denver, CO, 1997.
2. Zamansky, V.M., Ho, L., Maly, P.M., and Seeker, W.R. *26th Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA, pp. 2075-2082, 1996.
3. Zamansky, V.M., Sheldon, M.S. and Maly, P.M. *27th Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA, pp. 2001-2008, 1999.
4. Mungal, M., Han, D., Hasselbring, E., Lutz, A., Broadwell, J., Tyson, T., Zamansky, V., Serauskas, R., and Kezerle, J. *Presented at the 1998 International Gas Research Conference*, San Diego, CA, Paper IPP-25, 1998.
5. Xu, H., Smoot, L.D., and Hill, S.C. *Energy and Fuels* 13:411-420 (1999).
6. Chen, S.L., Cole, J.A., Heap, M.P., Kramlich, J.C., McCarthy, J.M., and Pershing, D.W., *22nd Symposium (International) on Combustion*, The Combustion Institute, Pittsburgh, PA, pp. 1135-1145, 1988.
7. Zamansky, V.M., Ho, L., Maly, P.M., and Seeker, W.R., *The Fourth International Conference on Technologies and Combustion for a Clean Environment, Section 10.1*, Lisbon, Portugal, pp. 1-9, 1997.
8. Alzueta, M.U., Røjel, H., Kristensen, P. G., Glarborg, P., and Dam-Johansen, K., *Energy and Fuels* 11:716-723 (1997).
9. Zamansky, V.M. and Lissianski, V.V., *Israel Journal of Chemistry* 39:63-71 (1999).

10. Kau, C. J., Heap, M. P., Seeker, W. R., and Tyson, T. J., Fundamental Combustion Research Applied to Pollution Formation. *U.S. Environmental Protection Agency Report No. EPA-6000/7-87-027, Volume IV: Engineering Analysis*, 1987.
11. Kee, R.J., Rupley, F.M. and Miller, J.A., Chemkin II: a Fortran Chemical Kinetics Package for the Analysis of Gas Phase Chemical Kinetics, *Sandia National Laboratories Report SAND89-8009*, 1992.
12. Glarborg, P., Alzueta, M.U., Dam-Johansen, K., and Miller, J.A., *Combust. Flame* 115:1-27 (1998).
13. Cetegen, B. M., Johnson, T. R., Payne, R., Moyeda, D. K., and Sheldon, M. S., Effective Mixing Processes for  $\text{SO}_x$ , Sorbent, and Coal Combustion Products, *U.S. Environmental Protection Agency Report No. EPA/600/7-87/013*, 1987.
14. FLUENT, *Fluent 5 User's Guide*, Fluent, Inc., Lebanon, NH, 1998.

### Figure Captions

Figure 1. Evaporation time as a function of droplet diameter.

Figure 2. Comparison of modeling predictions (lines) with experimental data (symbols) on the effect of temperature at the point of OFA/N-agent injection on  $\text{NO}_x$  reduction at 2% (open circles) and 10% reburning (filled circles). NSR=1.5.

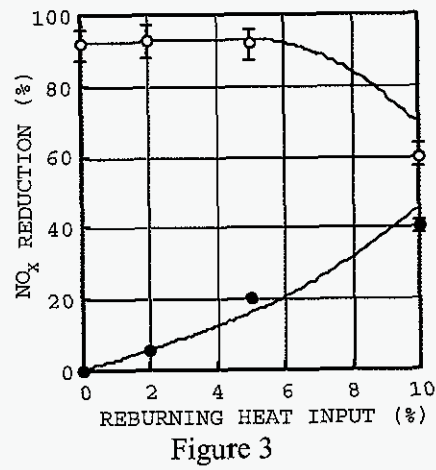
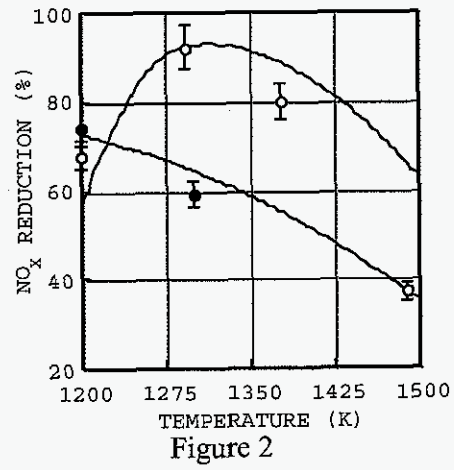
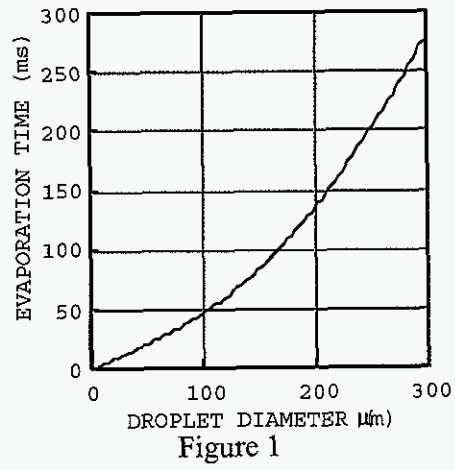
Figure 3. Comparison of modeling predictions (lines) with experimental data (symbols) on basic reburning (filled circles) and AR-Lean reburning (open circles). NSR=1.5. OFA and N-agent are injected at 1300 K.

Figure 4. Performance of the AR-Lean process at NSR=1.5 (a) and NSR=0.7 (b). Lines represent calculations, symbols experimental data. Numbers indicate levels of  $\text{NO}_x$  reduction. Evaporation time of the N-agent is less than OFA mixing time.

Figure 5. Performance of AR-Lean at NSR=0.7 as a function of the amount of the reburning fuel and droplet evaporation time of N-agent. Numbers indicate levels of  $\text{NO}_x$  reduction. Temperature of flue gas at the point of OFA/N-agent injection is optimized.

Figure 6. Predicted performances of basic reburning, AR-Lean and Reburning+SNCR. 1 – basic reburning, 2 – Reburning+SNCR, 3 – AR-Lean optimized, 4 – AR-Lean non-optimized. NSR=0.7.

# Figures



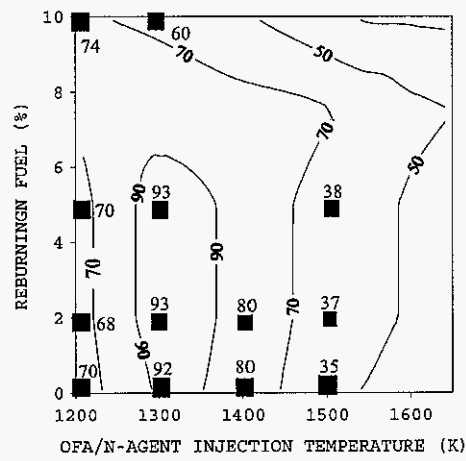


Figure 4a

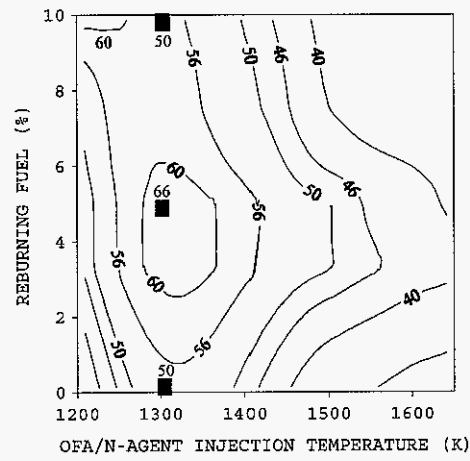


Figure 4b

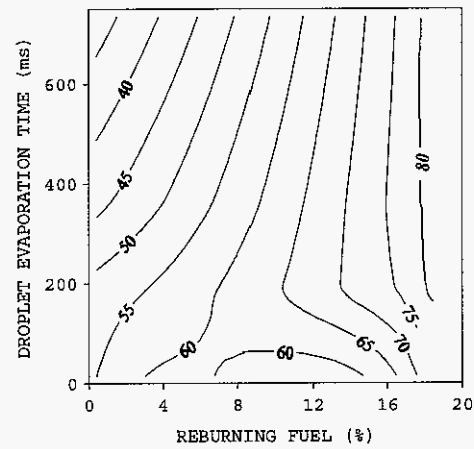


Figure 5

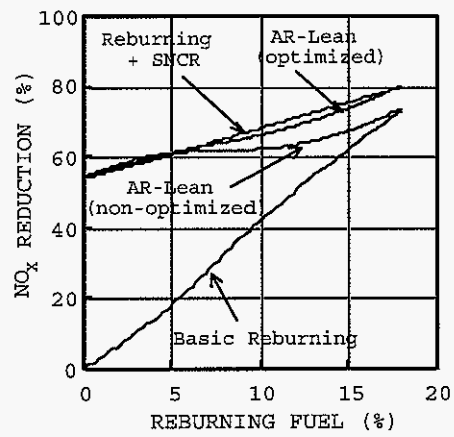


Figure 6